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CRITERIA FOR SELECTING RESIN MATRICES FOR IMPROVED COMPOSITE STRENGTH

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ABSTRACT

Criteria are presented which can be used a priori to select matrices to yield composites with improved unidirectional strength. The criteria evolved from an investigation including both theoretical and experimental efforts. Composite micromechanics were used in conjunction with suitable experiments and reference data to identify those matrix properties which influence composite strength properties. Composites of graphite fibers and low, intermediate and high-modulus resins were investigated. It was found that the area under the matrix stress/strain diagram limited to one percent strain is a good index for an a priori assessment of the matrix contribution to composite strength. The corresponding initial modulus is a useful parameter in translating matrix properties to composite properties. Matrix properties such as ultimate strength, ultimate elongation, toughness, and fracture toughness are misleading in translating matrix properties to composite strength.

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I - INTRODUCTION

There is a large number of resin matrices available which can be used to make fiber reinforced plastic composites. These matrices have properties differing in modulus, toughness, elongation, strength, and strength retention at elevated temperatures. In addition, the mechanical properties of any one matrix can be substantially altered by suitable additives. The researcher and/or a designer is confronted with the problem of a priori selecting matrices from the large number of available matrices which will yield composites with improved mechanical properties. The main difficulty for the selection of a matrix resin is that matrix mechanical properties do not transfer to composite mechanical properties in a parallel correspondent manner.

Considerable effort has been expended in the fiber composite community to correlate matrix properties with composite strength. In recent years, these efforts have concentrated on correlating composite strength with matrix ultimate strength (ref. 1), ultimate elongation (ref. 2), fracture toughness (refs. 3 and 4), and other matrix properties (refs. 1, 2, 5, 6, and 7).

Taken individually or jointly, no criterion has emerged from the aforementioned efforts which can be used to select matrices a priori.

The reasons for this are as follows: (1) the investigations concentrated on ultimate matrix properties, (2) they focused mainly on a single matrix property to correlate with one or more composite unidirectional strengths, (3) the investigations did not include micromechanics considerations for all the composite unidirectional strengths in conjunction with a parallel experimental effort.

It was suggested in references 2 and 8 that perhaps the matrix properties/composite strength correlation is governed by some combination of matrix properties. It was pointed out in references 9 and 10 that: (1) the in situ resin matrix is not stressed to its ultimate strength or elongation at the onset of composite failure and (2) only a small portion of the matrix stress strain diagram was working with the composite. These observations and the absence of a criterion for the a priori selection of resin matrices for improved composite strength lead to the present investigation.

The objective of the present investigation was to evolve convenient and practical criteria which can be used to select matrices which will yield composites with improved unidirectional strengths. The investigation is a combination of theoretical and experimental efforts. Composite micromechanics are used in conjunction with experimental data generated in this investigation and taken from the literature to identify those matrix properties which influence composite unidirectional strength. Unidirectional specimens were made from graphite fibers and low-modulus, intermediate-modulus, and high-modulus resins. These specimens were tested for strength in longitudinal flexure, transverse tension, and compression, and interlaminar shear. The corresponding available composite strength data (ref. 7) included longitudinal tension and compression, transverse tension, and intralaminar shear. Comparisons of composite strength data with the various matrix properties led to the identification of convenient and practical criteria which can be used to assess a priori the matrix contribution to composite strength. These criteria can also be used to guide polymer researchers to develop new matrices. A byproduct of the investigation was to suggest a standard procedure for determining initial matrix modulus.

II - THEORETICAL CONSIDERATIONS FOR IDENTIFYING MATRIX PROPERTIES INFLUENCING COMPOSITE STRENGTH

The theoretical portion of this investigation is concerned with some fundamental physical phenomena and with a brief review of micromechanics strength theories. The discussion on the micromechanics strength theories is limited to two main considerations: (1) a summary of the available equations for predicting composite uniaxial strengths from constituents, and (2) identification of the important matrix variables in these equations.

A - Fundamental Concepts

A schematic of the unidirectional composite of interest in this investigation is shown in figure 1. The uniaxial strengths noted in figure 1 are: longitudinal tension ($S_{\ell 11T}$), longitudinal compression ($S_{\ell 11C}$), transverse tension ($S_{\ell 22T}$), transverse compression ($S_{\ell 22C}$), and intralaminar shear ($S_{\ell 12S}$) as measured by a thin tube subjected to torsion. Interlaminar shear, as used herein, refers to shear values measured in a short beam shear test.

Experimental data (refs. 11 and 12) show that practically all unidirectional fiber/resin composites loaded in longitudinal tension exhibit linear stress/strain relationship to fracture. Two possibilities exist for the corresponding relationships of the constituent materials. These are: (1) both linear, or nearly so, as illustrated in figure 2; or (2) linear for the fiber and nonlinear for the matrix as shown in figure 3. The first possibility is predominant in advanced fiber/resin composites. The second occurs mainly in metal matrix fiber composites (ref. 13). It may also occur in advanced fiber/resin composites subjected to an environment which degrades matrix properties, such as elevated temperature. By elevated temperature is meant a temperature close to the composite cure temperature or glass transition temperature of the resin.

B - Matrix Properties Influencing Composite

Longitudinal Tensile Strength

The expressions most often used to predict longitudinal tensile strength are based on either the rule-of-mixtures or on statistical considerations. The modified rule-of-mixtures expression is given by (ref. 10).

$$S_{\ell 11T} = S_{fT}\left(\beta_{fT}k_f + \beta_{mT}k_m \frac{E_{m11}}{E_{f11}}\right) \tag{1}$$

where $S_{\ell 11T}$ is the composite longitudinal strength; β_{fT} and β_{mT} are theory/experiment correlation coefficients; E_{f11} and E_{m11} are longitudinal fiber and matrix moduli, respectively; S_{fT} is the fiber fracture stress, and k_f is the fiber volume ratio.

The fiber fracture stress is length dependent, figure 4. In equation form, S_{fT} is given by

$$S_{fT} = F(L_C) \tag{2}$$

where L_C is the fiber "critical length." The fiber "critical length" is defined as the minimum embedded-in-the-matrix length which will support sufficient stress to fracture the fiber (ref. 14). An estimate of the critical length can be obtained from the following equation which was derived using shear-lag-theory:

$$\left(\frac{L_C}{d_f}\right) = \frac{1}{2}\left(\frac{E_{f11}}{G_m}\right)^{1/2}\left[\left(\frac{\pi}{4k_f}\right)^{1/2} - 1\right]^{1/2} \ln(1 - c) \tag{3}$$

where d_f is the fiber diameter; G_m is the matrix shear modulus which is given by $E_m/2(1 + \nu_m)$; and c is the fiber stress transfer coefficient usually taken as $0.90 < c < 1.0$. See also reference 14.

The graphical representation of equation (3) is shown in figure 5 for S-glass/epoxy composites and in figure 6 for composites in general. Both of these figures show a strong dependence of L_C on matrix modulus.

The expression for $S_{\ell 11T}$ which can be derived using statistical considerations is given by (refs. 14 and 15):

$$S_{\ell 11T} = k_f(\alpha\beta L_C e)^{-1/\beta} \tag{4}$$

where α and β are the Weibull distribution function parameters for fiber strength, e is the natural logarithm, and L_C is given by equation (3).

It may be seen that both expressions for $S_{\ell 11T}$, equations (1) and (4), exhibit a dependence on L_C. It also may be seen that the only matrix properties in equations (1) and (3) are the matrix shear modulus and tensile modulus. Therefore, it may be concluded that the only matrix properties which influence $S_{\ell 11T}$ are the matrix moduli. Because the ratio E_m/E_f can range from 0.1 to 0.01 (for nearly all matrices and reinforcing fibers), the sensitivity of $S_{\ell 11T}$ to matrix modulus can vary within a wide range.

C - Matrix Properties Influencing Composite Transverse Tensile and Compressive and Intralaminar Shear Strengths

1. Governing equations based on matrix limit strain. - Stress-strain curves for high and low modulus matrix resins are shown in figures 7(a) and (b), respectively. Also included in the figures is the stress-strain curve for a unidirectional composite tested in the transverse direction. It can be seen from figure 7 that only the initial portion of the matrix stress/strain curve is utilized in the composite. The notation to be used in subsequent discussion is defined in figure 7. Note that the matrix limit strain, ε_{mpT}, is taken to be the point at which the matrix stress/strain curve exhibits a pronounced nonlinearity. A more useful definition will be given later.

The governing micromechanics equations are from reference 10:

Transverse tensile strength ($S_{\ell 22T}$)

$$S_{\ell 22T} = \beta_{22T} \frac{\varepsilon_{mpT}}{\beta_v \phi_{\mu 22}} E_{\ell 22} \tag{5}$$

Transverse compressive strength ($S_{\ell 22C}$)

$$S_{\ell 22C} = \beta_{22C} \frac{\varepsilon_{mpC}}{\beta_v \phi_{\mu 22}} E_{\ell 22} \tag{6}$$

Intralaminar shear strength ($S_{\ell 12S}$)

$$S_{\ell 12S} = \beta_{12} \frac{\varepsilon_{mpS}}{\beta_v \phi_{\mu 12}} G_{\ell 12} \tag{7}$$

The undefined notation in equations (5), (6), and (7) is as follows: β denotes the theory experiment correlation coefficient reflecting the fabrication process; β_v denotes the void influence; ϕ_μ is the matrix-strain-magnification factor; the subscripts T, C, and S denote tension, compression, and shear, respectively; ε_{mpT} is the matrix limit strain as defined from figure 7; and correspondingly for compression and shear; $E_{\ell 22}$ and $G_{\ell 12}$ are the composite transverse and shear moduli, respectively.

There are three groups of variables with distinct physical meaning in equations (5), (6), and (7). These groups can be easily identified by writing equation (5) in the following form:

$$S_{\ell 22T} = \left(\frac{\beta_{22T}}{\beta_v}\right)\left(\frac{E_{\ell 22}}{\phi_{\mu 22}}\right)\varepsilon_{mpT} \tag{5a}$$

where (β_{22T}/β_v) represents the particular fabrication process and depends only on the fabrication process; $(E_{\ell 22}/\phi_{\mu 22})$ is defined herein as the "strength parameter" which depends on the local and average composite geometry and on the elastic properties of the constituents (ref. 10); and ε_{mpT} is the matrix limiting strain as defined previously. Corresponding variables in equations (6) and (7) can be grouped in the same fashion with analogous physical interpretations.

The matrix variables influencing $S_{\ell 22T}$ enter through either $(E_{\ell 22}/\phi_{\mu 22})$ or ε_{mpT}. The group (β_{22T}/β_v) does not depend (at least not explicitly) on the matrix elastic or strength properties (ref. 10).

The variation of $(E_{\ell 22}/\phi_{\mu 22})$ and $(G_{\ell 12}/\phi_{\mu 12})$ with matrix modulus for a Thornel-50/epoxy composite with a 0.5 fiber volume fraction and zero voids is shown in figures 8 and 9, respectively. As can be seen in figures 8 and 9 the matrix modulus markedly affects the transverse and shear strength parameters.

The results in figures 8 and 9 suggest that transverse and intralaminar shear strength tests should be quite sensitive to matrix modulus.

The variation of the transverse strength parameter $(E_{\ell 22}/\phi_{\mu 22})$ with fiber volume ratio is shown in figure 10 for three matrix moduli, zero voids and 10 percent voids. The curves in figure 10 show that the transverse strength parameter is sensitive to both matrix modulus and void content. However, it is not sensitive to fiber volume ratio. These observations also apply to the intralaminar shear strength parameter.

2. Governing parameters considering possible notch sensitivity. - Experimental tensile tests show that epoxies exhibit brittle-type fracture (refs. 7 and 15). This implies that test specimens for transverse tensile strength can be notch sensitive resulting in a wide data scatter band. An assessment of the notch sensitivity of transverse tensile strength is obtained by using $\varepsilon_{mpT} = S_{mTI}/E_{m22}$ in equation (5a) which yields

$$S_{\ell 22T} = \left(\frac{\beta_{22T}}{\beta_v}\right)\left(\frac{E_{\ell 22}}{\phi_{\mu 22}}\right)\left(\frac{S_{mTI}}{E_{m22}}\right) \tag{5b}$$

where S_{mTI} is the in situ matrix tensile strength. Equation (5b) can be normalized with respect to S_{mTI}. The result is

$$\left(\frac{S_{\ell 22T}}{S_{mTI}}\right) = \left(\frac{\beta_{22T}}{\beta_v}\right)\left(\frac{E_{\ell 22}}{\phi_{\mu 22}}\right)\frac{1}{E_{m22}} \tag{5c}$$

$$S_{mTI} = K_{I_C mI}\Big/\sqrt{\pi a} \tag{5d}$$

where $K_{I_C mI}$ is the in situ matrix fracture toughness in the opening mode, and a is the crack length. The graphical representation of equation (5c) as a function of fiber volume ratio is shown in figure 11. The effects of matrix modulus and void content are also shown in this figure. Similar trends are obtained for intralaminar shear since equations (5) and (7) have the same form.

It is clear from the results shown in figure 11 that the transverse strength parameter $S_{\ell 22T}/S_{mTI}$ is insensitive to matrix modulus, insensitive to fiber volume ratio, but sensitive to void content. These observations lead to the following implications:

(1) The composite transverse tensile strength is probably governed by the fracture-mechanics opening mode (mode I).

(2) The composite transverse compressive strength is probably governed by the fracture-mechanics in plane shear mode (mode II). Specimens tested in transverse compression fail at 45° to the load direction as will be described in the experimental section.

(3) The conventional narrow specimens are not suitable for determining the matrix modulus effect on the transverse tensile or compressive strengths.

It is important to note that the horizontal-type-beam shear specimen (interlaminar shear) is not notch sensitive because this specimen is subjected to complex loading. It is reasonable to expect that the tubes and torsional rods subjected to shear will be more sensitive than the horizontal-beam-shear. It is anticipated that any of these tests will be a sensitive test to assess matrix modulus effects.

D - Matrix Properties Influencing Composite

Longitudinal Compressive Strength

Several equations have been proposed to predict longitudinal compressive strength. Herein, only four are examined. These are:

Modified rule of mixtures (ref. 10):

$$S_{\ell 11C} = S_{mC}\left(\beta_{mC}k_f + \beta_{fC}\frac{E_{f11}}{E_{m11}}\right) \tag{8}$$

Symmetric microbuckling (ref. 14):

$$S_{\ell 11C} = 2k_f\left[\frac{k_f E_{m11} E_{f11}}{3(1 - k_f)}\right]^{1/2} \tag{9}$$

Nonsymmetric microbuckling (ref. 15) or panel microbuckling (refs. 16 and 17):

$$S_{\ell 11C} = \frac{G_{m12}}{1 - k_f} \tag{10}$$

Debonding-intralaminar shear (ref. 10):

$$S_{\ell 11C} = a_1 S_{\ell 12S} + a_2 \tag{11}$$

The undefined notation in equations (8) through (11) is as follows: The subscript C denotes compressive property. The variable S_{mC} is the matrix-compressive-limit stress measured at a point on that matrix compressive stress/strain curve corresponding to ε_{mp} in figure 3. The intralaminar shear stress $S_{\ell 12S}$ is given by equation (7). The parameters a_1 and a_2 are evaluated as described in reference 10 or 18.

The dependence of $S_{\ell 12S}$ on matrix modulus is shown in figure 9 through the strength parameter $G_{\ell 12}/\phi_{\mu 12}$. Therefore it is concluded from equations (8) to (11) that the longitudinal compressive strength depends on matrix compressive strength and matrix modulus. Longitudinal compressive tests should be sensitive tests to assess the influence of the matrix modulus on $S_{\ell 11C}$. This test will not be as sensitive if longitudinal compressive failure is not caused by either microbuckling or intralaminar shear.

E - Matrix Properties Influencing Composite

Longitudinal Flexural Strength

The test for longitudinal flexural strength subjects the specimen to a combined stress state. A three-point bending test specimen and its stress distribution are illustrated in figure 12. As can be seen in figure 12, longitudinal tension, longitudinal compression or interlaminar shear could initiate fracture individually or in various combinations. The following criteria can be used to determine which stress would be the first to reach its critical value and cause fracture. These criteria are:

Longitudinal tensile fracture mode:

$$\sigma_{\ell 11} \geq S_{\ell 11T} \tag{12}$$

Longitudinal compressive fracture mode:

$$\sigma_{\ell 11} \leq -S_{\ell 11C} \tag{13}$$

Interlaminar shear fracture mode:

$$\sigma_{\ell 12} \geq S_{\ell 12S} \tag{14}$$

where it was assumed that the interlaminar shear strength may be approximated with $S_{\ell 12S}$, figure 12.

The variation of the strengths $S_{\ell 11T}$, $S_{\ell 11C}$, and $S_{\ell 12S}$ with matrix modulus has been demonstrated previously. Here, it merely needs to be pointed out that if fracture were caused by condition (12), the matrix contribution would be insignificant. If fracture were initiated by condition (13), then it would be caused by either microbuckling or intralaminar shear and the matrix contribution would be significant. If fracture were initiated by condition (14), then it would be essentially a short-beam-shear test and the matrix contribution would, of course, be significant. The conclusion, therefore, is that the flexural test should be a sensitive test to assess matrix contribution. The test will not be as sensitive if fracture is initiated by tension. The bending test should be a sensitive test in environments which are suspected to degrade or improve matrix properties.

An additional point that can be made based on the previous discussion and figure (12) is as follows: Tensile or compressive strengths measured from flexural tests are usually higher than those obtained from uniaxial tests because in a flexural test specimen the longitudinal stress distribution (fig. 12(b)) does not remain linear as the fracture progresses.

F - Possible Sensitive Tests for Assessing Matrix Influence on Composite Strength

The micromechanics considerations described previously are summarized in table I. The predominant matrix property influencing the particular composite strength is indicated. Recommended test methods, type of sensitivity anticipated and comments pertinent to each composite-strength/matrix-property/test-method are also given.

In cases where more than one matrix property influence composite strength, for example transverse tensile, the area under the initial portion of the matrix stress/strain curve is probably a better index of matrix contribution.

III - EXPERIMENTAL INVESTIGATION

The experimental part of the investigation is described in this section. The description consists of material selection, specimen fabrication, test apparatus and procedure, and results obtained and discussion.

A - Materials Selection

The constituent materials selection was based on their properties which strongly influence composite strength as predicted by the micromechanics. Two continuous graphite fiber materials were used, (1) HT-S tow and (2) Thornel 50S yarn. Both fibers were surface treated by the fiber manufacturers to improve intralaminar shear. Three resin systems were selected on the basis of tensile modulus.

B - Materials and Specimen Fabrication

Cast epoxy resins as well as graphite fibers in combination with epoxy matrices were evaluated in the experimental investigation. The resins and fibers used are listed in table II. The cast resins were obtained by molding sheets of 0.125-inch thickness. The resins were polymerized using cure cycles recommended by the manufacturer. Tensile specimens were machined in accordance with ASTM standard method D638 - type 1.

Fiber/resin composites were prepared from fiber that was drum wound and impregnated with the resin. The required number of unidirectional plies were molded in a 3 × 10-inch matched-die mold to produce the desired laminate thickness. The fiber direction was in the 10-inch dimension. Transverse tensile and compression test specimens were machined from 0.50 × 3.0-inch by 0.20-inch thick composite coupons. The specimen configuration was determined by a template which was used during the machining operation. The tensile specimens had a reduced test section of 0.25-inch wide by 1.5-inch long. The ends of the specimens were provided with metal-reinforced holes for pinned-loading fixtures. The compression specimens were machined from coupons identical to those used for the transverse tensile specimens. The dimensions of this reduced test section were 0.25-inch wide by 0.75-inch long.

Flexure and short-beam interlaminar shear specimens were machined from 0.065-inch thick laminates. Flexure specimens were 0.50-inch wide by 0.065-inch thick laminates. Shear specimens were 0.25-inch wide by 0.50-inch long.

C - Test Apparatus and Procedure

All tests were performed in a universal testing machine with a selected constant-speed crosshead. Tensile tests of the cast resins were in accordance with ASTM standard method D 638. Tests were performed at a crosshead speed of 0.10-inch per minute. Strain to fracture was measured with a clamp-on extensometer.

In tests of composite materials, a crosshead speed of 0.05-inch per minute was used. Transverse-tensile specimens were loaded by means of pinned attachments to provide alinement in the tensile machine. Transverse-compression specimens were provided with end grips to provide support and load transfer during testing in a compression fixture.

The unidirectional flexure specimens were tested using the ASTM standard method D790-71-Method I. Tests were made on a three-point loading fixture having a span of 2.0-inch. The resulting span to thickness ratio was approximately 31:1. The short-beam interlaminar shear specimens were tested using a three-point loading fixture having a span to thickness ratio of 5:1.

D - Results and Discussion

The results obtained from the experimental program consist of stress/strain diagrams of the matrices investigated, photographs of fractured composite specimens, and composite properties.

The stress/strain diagrams of the matrices are shown in figure 13. As can be seen from this figure, the matrices selected differ considerably in initial and final properties.

Photographs of typical fractures of composite specimens are shown in figure 14. The specimens are from flexural, transverse, and interlaminar shear strength tests. Note that fracture initiated on the compression side of specimen with the low-modulus matrix and on the tensione side for the specimen with the high modulus matrix. This change of fracture initiation mode was expected and was discussed previously in the theoretical considerations (sections 11-F, C, also see table I).

The measured composite data are tabulated in table III for the two systems investigated. Because of variations in the fiber content of the various specimens, and for the purpose of comparison, the data are normalized to 50 percent fiber volume ratio. The data are based on tests of more than five specimens for each property determination. The results for the transverse tensile ($S_{\ell 22T}$) and compressive ($S_{\ell 22C}$) strengths given in table III indicate that these strengths are not sensitive to any individual matrix properties listed in the table. However, the other strengths are sensitive to one or more matrix properties.

Additional photographs and photomicrographs of fractured specimens are given in reference 7. One interesting observation from the photomicrographs given in reference 7 is that the fracture surfaces for longitudinal compression and intralaminar shear exhibit almost identical fracture modes and are indistinguishable in most instances. This is consistent with the intralaminar shear fracture mode predicted by equation (11) and with other available experimental data to be described later.

IV - IDENTIFICATION OF MATRIX PROPERTIES INFLUENCING COMPOSITE STRENGTH FROM EXPERIMENTAL DATA

The data generated from the experimental program, described in section III, and that from reference 7 are examined to identify matrix properties which influence composite strength. Also, other available experimental data which is pertinent to the present objective will be examined.

A - Data from the In-House Experimental Program

The data generated by the in-house experimental program is summarized in table III. The data for the HTS/epoxy composite is presented in a different format in table IV. The top portion of table IV contains the matrix properties data and the lower portion the composite data. Identification of the matrix property influencing composite strength is obtained in the following manner: Each row of composite strength data is compared with each row of the matrix data. The products of two matrix properties have been included with the matrix data. These products are:

(1) (Proportional-limit-stress) times (proportional-limit-strain)

(2) (Initial-modulus) times (fracture-toughness)

The first represents twice the area under the initial portion of the matrix stress/strain diagram and will be referred to as the "proportional-limit-area." The second evolved during the course of this investigation and could be an index for notch-sensitive strengths, such as transverse tensile.

In the comparisons which follow, the term "strongly influenced" denotes an increase in matrix property which produces a corresponding increase in composite strength. "Mild or slight influence" denotes an appreciable increase in matrix property which produces a relatively small increase in strength. "No influence," "inconclusive," or "adverse influence" are also used.

For example, comparison of the composite flexural strength row with the rows of matrix properties in table IV shows the following:

1. Flexural strength is strongly influenced by initial modulus, proportional limit stress, ultimate strength, and proportional limit area (initial area).

2. Flexural strength is mildly influenced by proportional limit strain.

3. Flexural strength is <u>not</u> influenced by toughness, fracture toughness, and the notch-sensitivity parameter.

4. Flexural strength is adversely influenced by the ultimate elongation.

Continuing the comparison for the other composite strengths in table IV it can be seen that:

1. The transverse tensile and compressive strengths are not influenced by any of the matrix properties listed in table IV. Comparisons with the notch-sensitivity parameter are inconclusive.

2. The interlaminar shear strength is strongly influenced by the modulus, proportional limit stress, ultimate strength, and initial area. It is mildly influenced by the proportional limit strain. It is not influenced by toughness, fracture toughness, and notch-sensitivity param-

eter. The intralaminar shear strength is adversely influenced by the ultimate elongation.

B - Data from Reference 7

Pertinent data from reference 7 is summarized in table V. The matrix properties are summarized in the top portion of the table and the composite strengths on the lower portion. As can be seen in table V, five resins were investigated.

Comparisons of the composite strengths with matrix properties is made in the manner described for the flexural strength section IV-A.

From the results shown in table V, it can be seen that:

1. The composite longitudinal tensile strength is strongly influenced by the matrix modulus. It is not influenced by the ultimate elongation and not influenced by the remaining properties.

2. The composite longitudinal compressive strength is mildly influenced by modulus, proportional limit stress and strain, and initial area. It is adversely influenced by the ultimate-elongation and not influenced by the remaining matrix properties.

3. The transverse tensile and intralaminar shear strengths increase mildly with matrix modulus.

In both the experimental program and the reference data, the observed results showed a strong influence of matrix modulus on composite strength as was anticipated from the theoretical considerations. The experimental data showed that some matrix properties are interrelated as would be intuitively expected.

C - Data from Other Sources

An extensive investigation on composite longitudinal compressive strength is reported in references 19 and 20. The results showed that compressive strength is strongly influenced by modulus in composites with soft matrices (matrix shear modulus less than 100 000 psi). Analogous results were reported in reference 21 for boron-epoxy composites.

In a survey paper on the status of nonmetallic matrix effects on composite properties (ref. 22) it is reported that interlaminar shear strength increases linearly with matrix ultimate tensile strength. This is consistent with micromechanics and experimental data discussed previously. It is also reported in reference 22 that both composite longitudinal compressive and flexural strengths increase linearly with interlaminar shear strength. Simple algebraic substitutions in equations (11) and (13) show that the longitudinal compressive and flexural strengths will increase linearly with interlaminar shear strength. This implies that these strengths are strongly influenced by the matrix tensile strength. No data on matrix modulus is reported.

Elevated temperature (130° C, 266° F) test data reported in reference 15 showed a 60 percent decrease in the initial matrix modulus from its room temperature value and an 80 percent decrease in matrix ultimate tensile strength. The corresponding decrease in composite longitudinal strength was 14 percent. These results show a strong influence of both matrix modulus and ultimate strength on longitudinal tensile strength.

Data obtained at cryogenic temperatures (ref. 23) showed the following:

1. The initial matrix modulus increased considerably.

2. The matrix ultimate tensile strength was not affected or decreased slightly.

3. The composite interlaminar shear strength markedly increased.

These results indicate a strong influence of matrix modulus on interlaminar shear strength and no influence of the matrix ultimate tensile strength.

In all of these examples, the composite strength was strongly influenced by the matrix modulus. Whereas, in some cases, composite strength was not influenced by matrix ultimate strength. In all instances, the observed results and/or trends were or would be anticipated from micromechanics considerations.

V - RECOMMENDATIONS

The results of this investigation lead to the two following recommendations: (1) criteria for selecting matrices for improved composite strength and (2) a proposed convenient procedure for determining the matrix initial modulus.

A - Criterion for Selecting Matrices to Yield Composites with Improved Properties

The previous discussion of theoretical and experimental results indicate that composite strength is sensitive to the following matrix properties: modulus, proportional limit stress and strain, and ultimate strength. The experimental data shows that these matrix properties are not independent. For example, an increase in the matrix initial modulus is followed by increases in proportional limit stress and strain, and in ultimate tensile strength. The converse is also true (fig. 13 and tables IV and V). The proportional limit area includes three properties and can serve as a combined index.

It was observed in reference 7 that the matrix ERLA 4617 gave the best balanced Modmor II/epoxy composite properties. This was also the conclusion of the in-house experimental program, table III. It is seen in figure 13 that ERLA 4617 has the largest proportional limit area of the three matrix stress/strain curves. See also tables IV and V.

In view of the above findings/observations, the following criterion is recommended:

> "Of the various simple matrix properties, the area under the matrix stress/strain curve up to the proportional limit strain (initial area) is the best index for assessing matrix influence on composite strength and overall composite structural behavior."

For composite strengths, which are notch-sensitive, the "notch-sensitivity" parameter appears to offer some promise. This parameter is defined herein as the product of initial modulus and the fracture toughness. Additional experimental data is needed to assess the utility of the notch-sensitivity parameter.

B - A Convenient Procedure for Determining the Initial Matrix Modulus

The importance of the initial matrix modulus on composite properties was illustrated in the preceding discussion. Determination of the initial modulus and the corresponding proportional limit is somewhat arbitrary.

It is observed from experimental data that advanced fibers are linear to fracture and have fracture strains of about 1 percent or less. Exceptions to this are S-glass and PRD-49 fibers which have fracture strains of about 4 percent and 2 percent, respectively, when tested along the fiber direction. The matrix stress/strain curves are almost linear in zero to one percent strain range as is readily observed in figure 13.

In view of these observations it is recommended that the proportional limit strain/stress and initial modulus be determined as follows:

1. The proportional limit strain/stress of the matrix stress/strain curve be taken as equal to one percent strain.

2. The initial modulus be taken as the secant modulus from the origin to the proportional limit strain point.

This concept is illustrated in figure 15 which is for tensile data. It is noted that stress/strain diagrams for shear and compression show similar behavior, reference 24. Comparing initial tangent and secant moduli values in figure 15, it is observed that the difference is negligible. An additional advantage of this concept is that values measured by various researchers can be easily compared. Though the concept was illustrated using tensile data, it is applicable to shear and compressive data as well.

VI - CONCLUSIONS

The results of this investigation lead to the following conclusions:

1. The area under the initial matrix stress/strain diagram and bounded by the one percent strain is a good index for an a priori assessment of the matrix contribution to composite strength and composite mechanical properties in general.

2. Using micromechanics strength theories, it is possible to identify matrix properties which influence composite strength and recommend possible test-methods to measure this influence. Also an assessment on the matrix-property/test-method sensitivity may be obtained.

3. Of the various matrix properties, matrix modulus is the governing parameter for the composite longitudinal tensile, compressive, and flexural strengths.

4. The composite intralaminar shear strength shows a strong dependence on either the matrix modulus or the matrix ultimate tensile strength.

5. The matrix ultimate strength, ultimate elongation, toughness, and fracture toughness are not suitable parameters to correlate composite strength with matrix properties.

APPENDIX - SYMBOLS

a_1, a_2	constants, eq. (11)
c	stress transfer coefficient, eq. (3)
d	diameter
E	longitudinal modulus
F	failure function
G	shear modulus
k	apparent volume ratio
L_C	critical fiber length for stress transfer
S	simple strength, failure or limit stress
t	thickness
β	theory-experiment correlation factor
ε	strain
ν	Poisson's ratio
σ	stress
ϕ_μ	strain-magnification factor

Subscripts:

C	compression, critical length
D	debonding
F	flexure
f	filament property
I	in situ property
ℓ	ply property
m	matrix property
p	limiting property

S	shear
T	tension
v	void
1,2,3	material axes (the 1-axis coincides with the filament direction)

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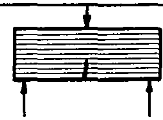
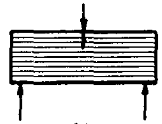
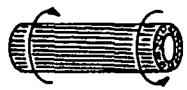
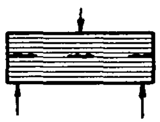
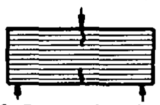
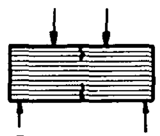
TABLE I. - MATRIX PROPERTIES INFLUENCING COMPOSITE STRENGTH, POSSIBLE TEST METHODS, AND SENSITIVITY

Composite strength	Predominant matrix properties	Recommended test method	Matrix properties sensitivity	Comments
Longitudinal tensile	Modulus	 Tensile  Bending	Slight	More sensitive for very soft matrices. Notch sensitive in composites with excessively high interlaminar shear strength
Longitudinal compression	Compressive strength, modulus	 Compressive  Bending	Good	Compression specimen should be prevented from gross buckling and end brooming. Flexure specimen sensitive for low modulus matrix and environmental effects
Transverse tensile	Modulus, limit strain, fracture toughness	 Tension	Slight	Notch sensitive. (Opening mode: Mode-I)
Transverse compressive	Modulus, limit strain, fracture toughness	 Compression	Slight	Notch sensitive. (Shear mode: Mode-II)
Intralaminar shear	Modulus, limit strain	 Torsion rod  Thin tube	Good	Fracture shows longitudinal split when the fibers are parallel to specimen length
Interlaminar shear	Modulus, limit strain	 Short-beam-bending	Excellent	Expedient for quality control and material comparative evaluation
Flexure (Bending)	Modulus	 3-Point bending  4-Point bending	Slight Good	If fracture initiated by tension. If fracture initiated by comparison.

General notes: (1) Design flexure specimens to avoid interlaminar shear failure. Report fracture initiation mode. Change in fracture mode will probably cause pronounced change in strength.
(2) Limit strain is measured at the point where pronounced nonlinearity begins on the respective stress/strain curve.
(3) In situ ply strength is probably a more suitable test for notch sensitive specimens.

TABLE II. - GRAPHITE/RESIN CONSTITUENT MATERIALS

Fibers	[a]HT-S, [b]Thornel 50S
Resin matrices	[c]ERLA 4617/mPDA/BF_3MEA (27.0, 1.5 PHR)
	[c]ERL 2256/ZZL 0820 (27.0 PHR)
	[c]ERL 2772/ZZL 0822 (20.0 PHR)

[a]Hercules Incorporated, Industrial Systems Department.
[b]Union Carbide Corporation, Carbon Products Division.
[c]Union Carbide Corporation, Chemicals and Plastics Division.

TABLE III. - MECHANICAL PROPERTIES OF CAST RESIN AND GRAPHITE COMPOSITES INVESTIGATED

Resin properties					Composite properties (50 percent fiber volume)				
Resin	Tensile strength,	Elastic modulus,	Elongation, percent	Impact energy, in.-lbs	Fiber	$S_{\ell 22T}$ psi	$S_{\ell 22C}$ psi	$S_{\ell 12S}$ psi	$S_{\ell 11F}$ psi
4617/mPDA	17.6×10^3	0.82×10^6	3.0	2.15	HTS	6.0×10^3	25.8×10^3	15.0×10^3	205×10^3
					T50S	4.5	----	7.4	121
2256/0820	14.0×10^3	0.56×10^6	4.3	3.07	HTS	6.3×10^3	25.6×10^3	11.5×10^3	150×10^3
					T50S	4.3	23.0	7.3	115
2272/0822	7.8×10^3	0.40×10^6	6.9	10.5	HTS	6.1×10^3	--------	7.8×10^3	122×10^3
					T50S	4.2	--------	6.1	105

TABLE IV. - SUMMARY OF MATRICES AND COMPOSITE PROPERTIES

HTS/EPOXY COMPOSITES

NASA LEWIS RESEARCH CENTER DATA

Property	Units	Resin		
		ERLA 2772	ERLA 2256	ERLA 4617
Matrix				
Initial modulus	10^6 psi	0.4	0.56	0.82
Proportional limit stress	10^3 psi	3.3	4.5	7.3
Proportional limit strain	percent	0.75	0.8	1.0
Ultimate strength	10^3 psi	7.8	14.0	17.6
Ultimate elongation	percent	6.9	4.3	3.0
Toughness = $\int_0^{\varepsilon_{ult}} \sigma \, d\varepsilon$	10^3 in.-lb/in.^{2}	0.417	0.368	0.310
Impact-strength	in.-lb	10.5	3.07	2.15
Proportional limit stress times proportional limit strain	in.-lb/in.^{3}	24.8	36.0	73.0
Initial modulus times impact strength	10^6 lb^2/in.	4.2	1.72	1.76
Composites (50 percent fiber volume)				
Composite longitudinal flexure strength ($S_{\ell 11F}$)	10^3 psi	[a]122.	[b]150.	[b]205.
Composite transverse tensile strength ($S_{\ell 22T}$)	10^3 psi	6.1	6.3	6.0
Composite transverse compressive strength ($S_{\ell 22C}$)	10^3 psi	---	25.6	25.8
Composite interlaminar shear strength ($S_{\ell 12S}$)	10^3 psi	7.8	11.5	15.0

[a]Compression failure.

[b]Tensile failure.

TABLE V. - SUMMARY OF MATRICES AND COMPOSITE PROPERTIES

MODMOR II/EPOXIES COMPOSITES (REF. 7)

Property	Units	Resin				
		ERLA 4289	EPON 828	ERLA 2256	ERLA 4617	ERLA 4305
Matrix						
Initial modulus	10^6 psi	0.24	0.32	0.50	0.78	0.89
Proportional limit stress	10^3 psi	2.2	3.6	5.0	8.6	7.5
Proportional limit strain	percent	0.3	1.2	1.0	1.3	0.9
Ultimate strength	10^3 psi	5.4	8.0	15.2	14.8	14.1
Ultimate elongation	percent	81.0	8.1	6.5	2.2	1.7
Toughness = $\int_0^{\varepsilon_{ult}} \sigma \, d\varepsilon$	10^3 in.-lb/in.^{3}	[a]0.35	0.91	0.71	0.18	0.14
Fracture toughness K_{IC} (center crack specimens)	$10^3 \frac{lb}{in.^2}$ in.$^{1/2}$	2.7	1.4	0.65	0.57	0.98
Proportional limit stress times proportional limit strain	in.-lb/in.^{3}	6.6	43.0	50.0	112.0	68.0
Initial modulus times fracture toughness	$10^9 \frac{lb^2 \, in.^{1/2}}{in.^4}$	0.65	0.45	0.33	0.44	0.87
Composites (Fiber volume ratio in parentheses)						
Composite longitudinal tensile strength ($S_{\ell 11T}$)	10^3 psi	116 [b](0.678)	205 (0.620)	207 (0.627)	150 (0.613)	231 (0.575)
Composite longitudinal compressive strength ($S_{\ell 11C}$)	10^3 psi	12.4 (0.673)	134 (0.614)	149 (0.598)	150 (0.671)	159 (0.664)
Composite transverse tensile strength ($S_{\ell 22T}$)	10^3 psi	1.2 (0.648)	3.4 (0.682)	3.9 (0.667)	4.3 (0.612)	2.9 (0.664)
Composite intralaminar shear strength ($S_{\ell 12S}$)	10^3 psi	0.77 (0.634)	3.7 (0.648)	6.9 (0.615)	7.9 (0.584)	6.1 (0.640)

[a]Measured at 10 percent strain.

[b]Fiber volume ratio.

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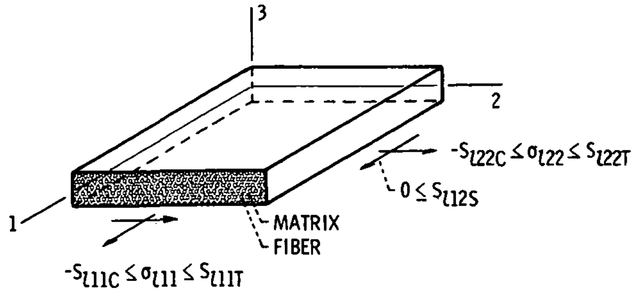

Figure 1. - Schematic of unidirectional composite showing geometry and uniaxial strength notation.

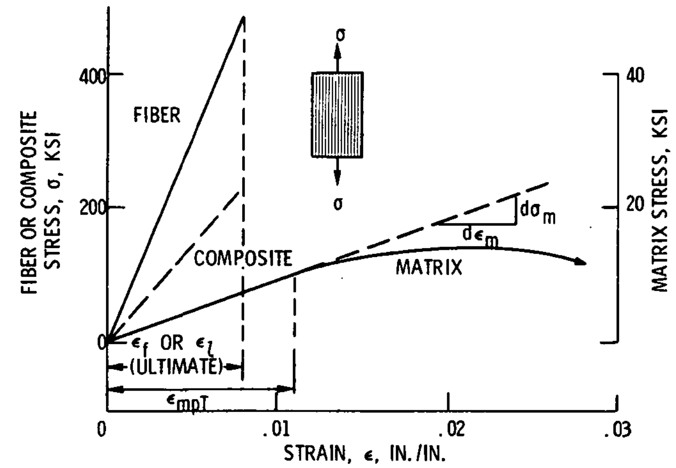

Figure 2. - Case where matrix remains linear or nearly so throughout the composite strain range.

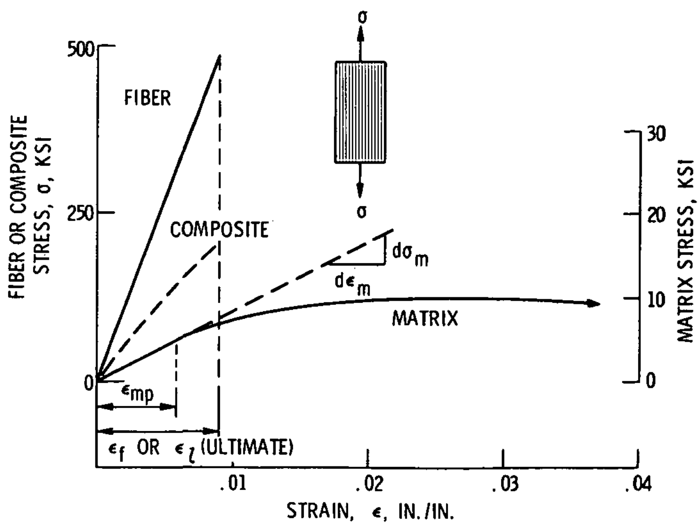

Figure 3. - Case where matrix behaves nonlinear in portion of the composite strain range.

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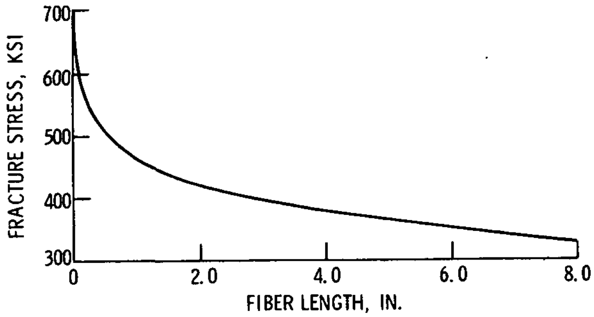

Figure 4. - Fiber gage length effect on fiber fracture stress S-glass fibers (ref. 24).

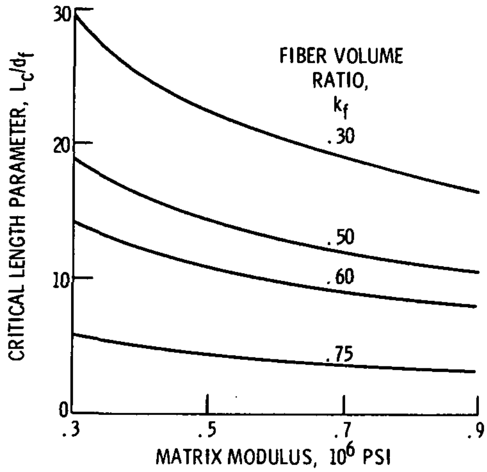

Figure 5. - Matrix modulus influence on critical length for S-glass/epoxy composites assuming 99.9 percent load transfer.

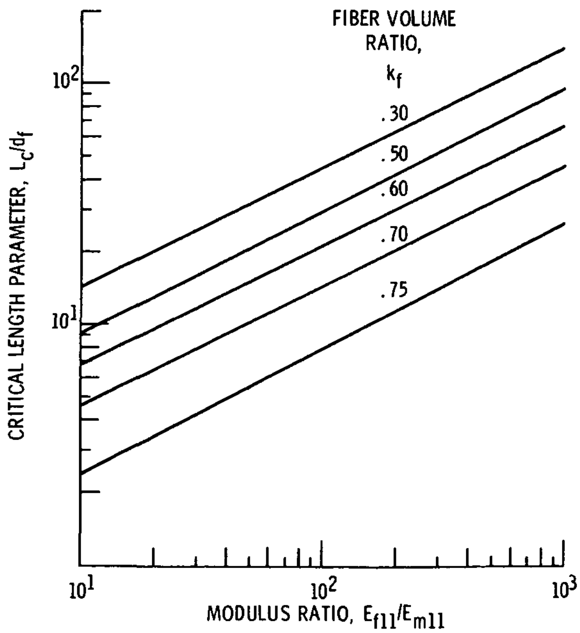

Figure 6. - Modulus ratio influence on critical length assuming 99.9 percent load transfer.

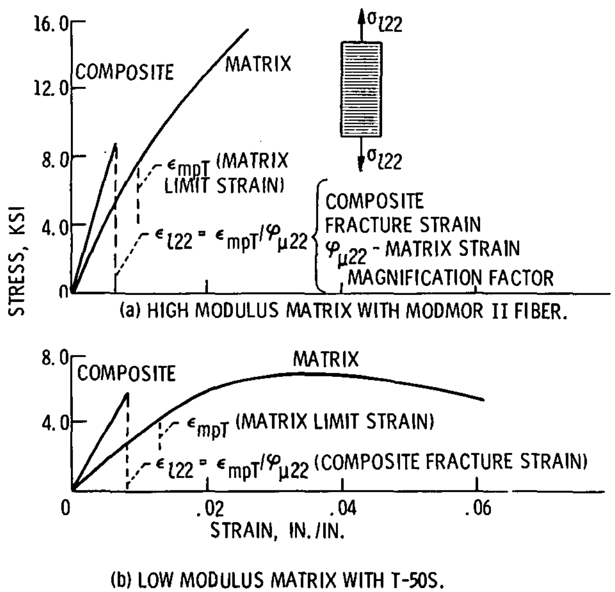

Figure 7. - Transverse composite and matrix stress/strain curves.

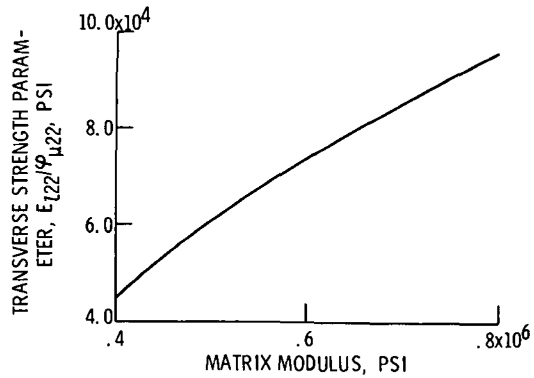

Figure 8. - Effect of matrix modulus on transverse strength parameter. TH-50/epoxy with 0.5 fiber volume ration and zero voids.

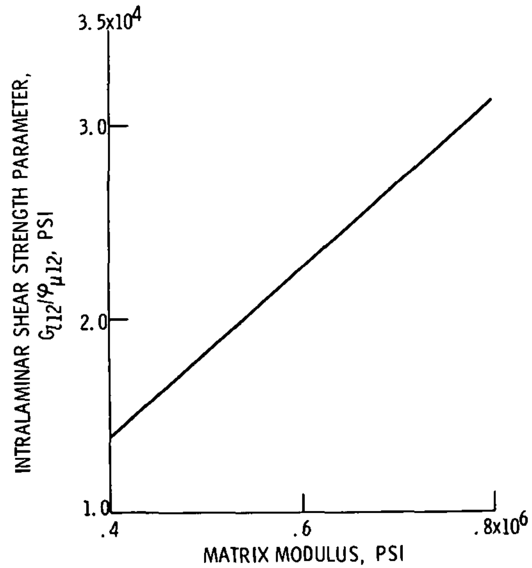

Figure 9. - Effect of matrix modulus on intralaminar shear strength parameter. TH-50/epoxy with 0.5 fiber volume ration and zero voids.

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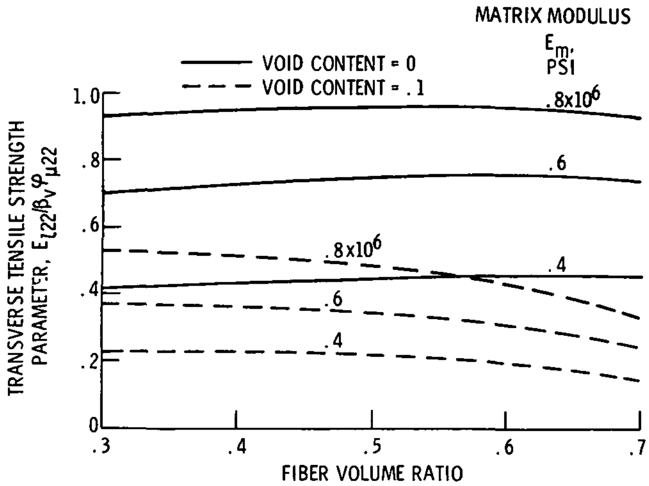


Figure 10. - Effects of Thornel-50/resin unidirectional composite transverse tensile strength limited by in situ matrix tensile strain (elongation).

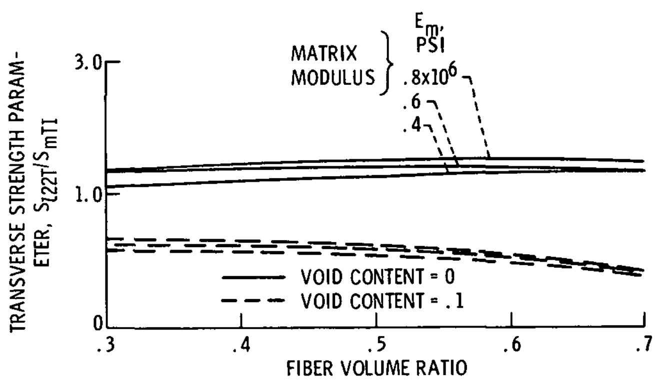
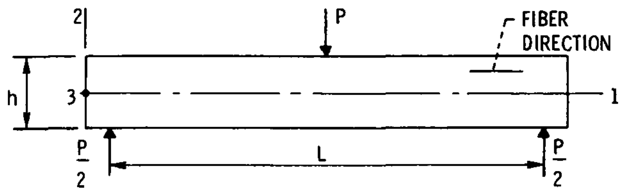
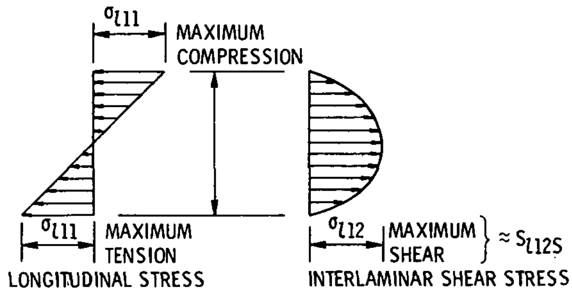


Figure 11. - Thornel-50/resin unidirectional composite transverse tensile strength limited by in situ matrix either tensile strength or fracture toughness.



(a) SPECIMEN GEOMETRY.



(b) STRESS DISTRIBUTION AT A SECTION NEAR THE LOAD.

Figure 12. - Geometry and stress distribution of 3-point bending test specimen.

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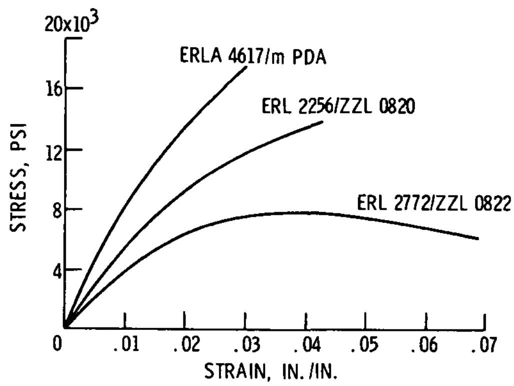
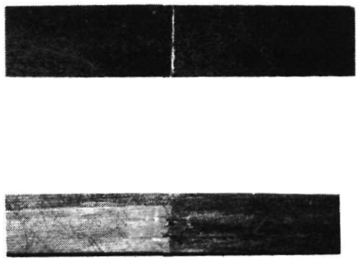


Figure 13. - Stress-strain diagrams of matrix resins.



COMPRESSION FAILURE
(LOW MODULUS MATRIX)

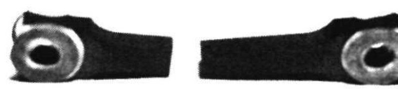
TENSILE FAILURE
(HIGH MODULUS MATRIX)

(a) UNIDIRECTIONAL FLEXURE.

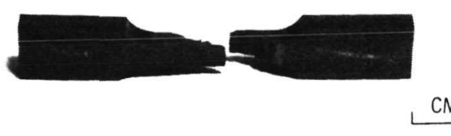
COMPRESSION FAILURE
(LOW MODULUS MATRIX)

TENSILE FAILURE
(HIGH MODULUS MATRIX)

(b) INTERLAMINAR SHEAR.



(c) TRANSVERSE TENSILE.



(d) TRANSVERSE COMPRESSION.

Figure 14. - Typical fractures of composite specimens subjected to various tests.

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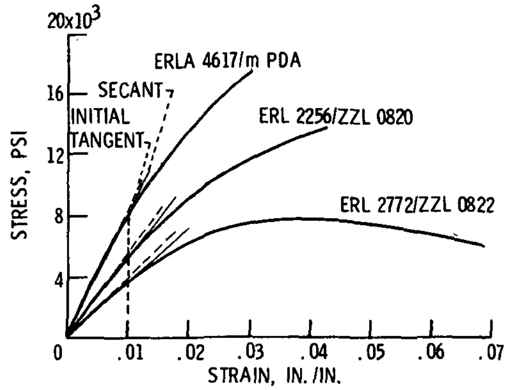

Figure 15. - Stress-strain diagrams of matrix resins with proposed definition for determining initial matrix modulus.